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Theoretical description of ABO_3 relaxor ferroelectric : A review

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Abstract : Significant similarities exist between spin glasses and relaxor ferroelectric. Because of the wealth of available knowledge both in experimental and theoretical aspects in the former, attempt has been made by many workers to deal the relaxor behavior in analogy with the spin glasses. However, the nature of diffused phase transition in relaxor ferroelectric remains controversial. To overcome this difficulty, many workers extended the above model to a new type of dipolar glasses, namely spherical vector glasses in which order parameter field is described as a continuous vector field of variable length instead of fixed length as proposed earlier. The present review is aimed to focus on all the models.

Keywords : Relaxor, ferroelectric, spin glass, ising model, SRBRF

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1. Introduction

Relaxor ferroelectrics, or relaxors exhibit many properties similar to those of spin or dipolar glasses. Relaxor behavior in normally ferroelectric materials results disorder or frustration [1]. This behavior has been observed and studied most extensively in disordered ABO_3 perovskite ferroelectric. Three essential ingredients of relaxor ferroelectric are the existence of lattice disorder, evidence of the existence of polar nanodomains at temperatures much higher than T_m and the existence of domains as islands in a highly polarisable host lattice. To close similarity, spin glasses are magnetic systems in which the interactions among the magnetic moments are both random and frustrated because of structural disorder. A common feature of these systems, and the one that makes them such an interesting object of study, is that they exhibit a freezing of magnetic moments in random directions at an apparently sharp temperature T_{sg} [2]. Because there are similarities between spin glasses and relaxor ferroelectric and

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because of the wealth of available knowledge in both experimental and theoretical front in the former, one can handle the relaxor behavior in analogy with the spin glasses. One can model the problem of relaxor ferroelectric in analogy with Ising model description of spin glasses.

2. Modeling

Under the most commonly studied Ising model of spin glass, consisting of N Ising spins S_i = ±1, i = 1, 2, ..., N, in the presence of an externally applied magnetic field H, the Hamiltonian for the model becomes [3],

$$H_H = -\frac{1}{2}\sum_{i,j} J_{ij} S_i \cdot S_j - H\sum_i S_i \ . \tag{1}$$

Here, each spin S_i interacts with every other spins S_j, with exchange transition J_{ij}. In term of Pauli spin matrices

$$H_H = -\frac{1}{2}\sum_{i<j} \boldsymbol{J}\left(\left|\boldsymbol{R}_i - \boldsymbol{R}_j\right|\right)\sigma_i\sigma_j - H\sum_i \sigma_i \tag{2}$$

where

$$\frac{1}{2} g\beta H_m = H \tag{3}$$

H_m being magnetic field strength.

Under the assumptions of one dimensional nearest neighbour interaction

$$H_H = -J\sum_n \sigma_n\sigma_{n+1} - H\sum_n \sigma_n \ . \tag{4}$$

Because the relaxors are materials with random site lattice disorder, it is natural to resort to models that investigate the role of random fields. In ABO_3 relaxors, the dipolar nanodomains formed by chemical substitutions create random electric fields in the host lattices. For an Ising spin model with infinite ranged interactions with statistically independent site field, one considers a collection of spin s = ±1 located at lattice sites i, j and interacting in such a away that the total energy, or Hamiltonion, is

$$H_H = -J\sum_{i\neq ij} S_i S_j - \sum_i \sigma_n \ . \tag{5}$$

Here the interaction J is a constant that incorporates all electronic properties in a phenomenological way, $<,ij>$. Designates nearest-neighbor pairs.

Although in eq. (5) the quantities S_{ij} are called “spins” the model is by no means restricted to magnetism. This model can describe any solid state system that has a transition with a doubly degenerate order state, and contains frozen impurities or point defects that break the symmetry and cannot move on the relevant time scale.

The important feature of relaxor is the presence of compositional and structural disorder, which breaks translation symmetry and produces random dipolar nanodomains. For relaxor as consisting of Ising like dipoles in a regular lattice with randomly disturbed exchange energy parameters, the Hamiltonian is simplified to

$$H_H = \sum_{i \neq j} J_{ij} S_i S_j + \frac{E\bar{\mu}\sum \mu_j \cos\theta}{\bar{\mu}} S_i \tag{5a}$$

where J_{ij} is assumed to have a Gaussian distribution with zero mean field value and a width Δj, μ_i is the magnitude of the dipole moment of the i-th polar nanodomains, θ is the angle between the external field E, and i-th dipole moment and μ is the maximum projection of the dipole moments on the main axis.

To find the free energy of this model as a function of temperature T, magnetic field H, and variance of random interactions σ, one can define partition function Z and introduce 2 × 2 matrices as under [3]

$$Z = \sum_{\sigma_1=\pm 1} \sum_{\sigma_2=\pm 1} ..... \sum_{\sigma_N=\pm 1} e^{H_H(\sigma_1 ..... \sigma_n)/k_B T} \tag{6}$$

$$(V_1)_{\sigma_i \sigma_j} = \exp\left(k_1 \sigma_i \sigma_j\right)$$

$$(V_2)_{\sigma_i \sigma_j} = \exp\left(H' \sigma_i\right) \delta_{\sigma_i \sigma_j} \tag{6a}$$

where $k_1 = J/K_B T$, $H' = H/K_B T$.

One can get the free energy per spin

$$F = \frac{-k_B T}{N} \log Z = -k_B T \log \lambda_1 \tag{7}$$

where

$$\lambda_1 = e^{k_1} \cos hH' + \sqrt{e^{2k_1} \sin h^2 H' + e^{-2k_1}} \ . \tag{8}$$

In the Monte Carlo simulation the polarization of this model is found to be

$$P = \frac{\int \langle \sigma(t) \rangle}{t_f - t_i} dt \tag{9}$$

where $<\sigma(t)>$ is the space averaged dipole at each Monte Carlo step.

The temperature dependence of the remnant polarization, P_R, can be calculated for experimental confirmation.

In spite of several attempts, the nature of diffused phase transition in relaxor ferroelectric has still remained controversial. Experimental evidence showed incompatibility with the assumptions of fixed length order parameter as proposed in dipolar glasses

or spin glasses. In the previous model we have described relaxors ferroelectric by an ordering field of fixed length under Ising model. Because of basic reorientable polar nanoclusters *i.e.* the "pseudospins" vary both in their size and orientation, the relaxor corresponds to new type of dipolar glasses namely spherical vector glasses and the order parameter field is described as a continuous vector field of variable length. Pseudospin Hamiltonian of a relaxor under Spherical Random Bond-Random Field (SRBRF) model takes the form [4].

$$H_s = -\frac{1}{2}\sum J_{ij}\boldsymbol{S}_i \cdot \boldsymbol{S}_j - \sum_i \boldsymbol{h}\cdot\boldsymbol{S}_i - g\boldsymbol{E}\cdot\sum_i \boldsymbol{S}_i \tag{10}$$

where $\boldsymbol{S}_i$-dipole moment of the i-th polar cluster, J_{ij}-random intercluster interactions (J_0/N -mean value, $(J)^2/N$ -variance), $\boldsymbol{E}$-external electric field, $\boldsymbol{h}_i$-random fields (0-mean value, $\left[h_{i\mu}h_{iv}\right]_{av}^{c} = \delta_{ij}\delta_{\mu v}\Delta$).

The model has two order parameters, the polarisation P and glass order parameter q. Equilibrium values of polarization $P = N^{-1}\sum_i \langle S_i \rangle$ and the glass order parameter $q = N^{-1}\sum_i \langle S_i^2 \rangle$ are determined from the condition of the free energy minimum. The knowledge of free energy per spin helps us to find polarisation and dielectric susceptibility. For isotropic system, spontaneous polarisation below T_c is given by [5],

$$P^2 = \left[1-\left(\frac{J}{J^0}\right)^2\right]\left[1-\left(\frac{T}{J^0}\right)-\frac{\Delta}{J^2}\right] \tag{11}$$

and the Edward-Anderson glass order parameter q_{EA} is given by

$$q_{\text{EA}} = 1 - T/j_0$$

where J_0 and J being the uncoupled interaction parameter and $\Delta = \left|J_0^2 - J^2\right|$.

Including the pseudospin lattice coupling interaction, for the nanodomains are being dispersed in a deformable lattice, the total Hamiltonian now becomes [4],

$$H = H_S + H_L + H_{SL} \tag{12}$$

where H_L and H_{SL} are the lattice (phonon) contribution and pseudospin polar phonon respectively. The lattice coupling modifies the uncoupled interaction parameters J_0 and J to J_0^* and J^* [6]. The phase transition to an inhomogeneous ferroelectric state occurs below transition temperature T_c given by

$$kT_c = J_0^*\left[1-\frac{\Delta}{(J_0^*)-(J^*)^2}\right]. \tag{13}$$

3. Conclusion

In summary, the coupled SRBRF-phonon model of relaxors appears to be successful in explaining the relaxor properties of ferroelectrics. Experimental work on relaxor ferroelectrics are in progress.

References

[1] G A Samara *Ferroelectrics* **117** 347 (1991) and references therein

[2] K Binder and A P Young *Rev. Mod. Phys*. **58** 801 (1986)

[3] J Callaway *Quantum Theory of Solid State Physics* (Academic Press) 2nd edition (1991)

[4] R Pric and R Blinc *Phys. Rev.* **B60** 13, 470 (1999)

[5] J J Van der Klink and F Borsa *Phys. Rev.* **B3** 52 (1984)

[6] G A Samara *Ferroelectric Revisited* (Advance in materials and Physics) (Academic Press) **56** 268 (2001)